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LETTER TO THE EDITOR

Interfacial correlation and dispersion in a non-equilibrium steady state system

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Abstract. Motivated by recent simulation results of the suppression of interfacial roughening, we study analytically the interfaces in non-equilibrium steady states for systems with bulk conservation. The general method of deriving interfacial properties from the bulk is developed, and the necessity of employing the full dynamics, even for static properties, is exhibited. Applying this to the randomly driven diffusion system, we obtained novel results. Specifically, height–height correlation diverges weakly as $1/q$ for small wavevector q, thus displaying the first analytical evidence of roughening suppression for driven systems.

Many interesting physical phenomena are primarily interfacial in origin and character, such as those of crystal growth (Langer 1987), nucleation and spinodal decomposition (Gunton *et al* 1984), roughening (van Beijeren and Nolden 1987), and membranes (Nelson *et al* 1989), to name just a few. They have attracted the attention of theoretical and experimental physicists for many decades. In addition, there is considerable interest in the relationship between interfacial and bulk degrees of freedom.

There exists a vast literature on interfaces, mostly devoted to systems at or close to equilibrium. By contrast, difficulties arising from the absence of a free energy hinder analyses of systems in non-equilibrium steady states, so that their properties are far less well explored and understood than their equilibrium counterparts.

Although there has been intense recent activity devoted to the physics of steadily growing interfaces, most studies are based on a phenomenological approach (e.g. Kardar *et al* 1985, Sun *et al* 1989, Derrida *et al* 1991), focusing on the interface variable alone, usually with local interactions. The advantage lies in its simplicity which, in some cases, even allows exact solutions. While this purely interfacial description is very useful, the neglect of bulk degrees of freedom can no longer be justified if the bulk dynamics is governed by conservation laws. For example, it is known that bulk diffusion induces non-local spatial interactions and memory effects for the interface (Langer and Turski 1977, Kawasaki and Ohta 1982). Such non-localities are responsible for the slower decay of interfacial fluctuations than those without bulk conservation. Therefore, it becomes necessary, for conserved systems, to derive interfacial properties by systematically eliminating the bulk degrees of freedom, starting from the full bulk equations.

Ideally, such an approach should be carried out for the entire microscopic system, by 'single out' the usually massive bulk modes. However, this turns out to be too difficult to be practical in most cases. Instead, for sufficiently simple systems, one normally starts, for statics, with a coarse-grained Landau–Ginzburg Hamiltonian (Diehl

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et al 1980), and for dyanmics (e.g. models A–D in critical dynamics), with the corresponding stochastic kinetic equations (Langer and Turski 1977, Bausch *et al* 1981, Kawasaki and Ohta 1982, Jasnow and Zia 1987, Bausch *et al* 1991).

The interplay between the interface and the bulk is even more important when the system is driven out of equilibrium. It has been recognized in recent years that correlations in driven systems often exhibit long-range, power-law decay (Spohn 1983, Bak *et al* 1987, Zhang *et al* 1988, Chen *et al* 1991). Conspiring with the usual non-local influences of bulk diffusion, such long-range effects inextricably couple the slow modes of the bulk to the interface. In addition, without the aid of an undisputed concept of free energy for non-equilibrium steady states, one must work with the dynamical equations, even for analysing time independent properties.

In this letter, we consider an interface separating the two coexisting phases far below criticality, in a system driven into a non-equilibrium steady state. A major motivation is the observation, in Monte Carlo simulations (Leung *et al* 1988, 1989), of the suppression of interfacial roughness in a diffusive lattice gas model, driven by a *uniform* external field E along an axis. So far, there is still no theoretical understanding of this effect. Here, we study a related but simpler model: the *randomly* driven system (Schmittmann and Zia 1991), in which the *magnitude* of E is a Gaussianly distributed, annealed random variable with zero mean. This system is 'simpler' because particle–hole symmetry is preserved and there is no global current. It is nevertheless a non-equilibrium system, displaying many of the characteristics of the uniformly driven case (Hwang *et al* 1991), e.g. a second-order phase transition at half-filled density. Our analysis shows that, e.g. interfacial correlations are indeed significantly suppressed, from the usual $1/q^2$ divergence to only $1/q$. Work is in progress to generalize this understanding to the uniformly driven case, in which the effect of roughness suppression was originally observed.

As revealed by computer simulations, the low-temperature configuration of the randomly driven system also consists of a particle-rich phase separated from particle-poor phase by a *planar* interface whose tangent is parallel to E. Since the prominent fluctuation is that associated with the interface, our main goal is the derivation of interfacial properties in the low-frequency, long-wavelength limit, from the bulk equation of motion.

Starting with the Langevin equation describing the action of $E(x, t)$ on ϕ (in the language of Ising spins, ϕ is the local spin density), we integrate out E to get new effective parameters. The resulting equation takes the form of a generalized model-B equation (Schmittmann and Zia 1991):

$$\frac{\partial \phi}{\partial t} = [r_{\|}\partial^2 + r_{\perp}\nabla^2 - (\alpha\partial^4 + \beta\nabla^4 + 2\gamma\partial^2\nabla^2)]\phi + (g_{\|}\partial^2 + g_{\perp}\nabla^2)\phi^3/3! + \eta \quad (1)$$

where $\partial(\nabla)$ stands for gradient in the parallel (transverse) direction with respect to E. For generality, we have allowed for full anisotropies, η is the Gaussian noise, correlated by $\langle\eta(x, t)\eta(x', t')\rangle = N\delta(x - x', t - t')$, with strength $N \equiv -(n_{\|}\partial^2 + n_{\perp}\nabla^2)$. This equation has the following important properties.

1. As a result of random stirring, the effective temperature is higher for the direction parallel to E (Schmittmann and Zia 1991, Cheng *et al* 1991). To model this effect, we assume $r_{\|} > r_{\perp} > 0$ above the critical temperature T_c. Thus, as $T \to T_c$ from above, $r_{\perp}$ vanishes first, while $r_{\|}$ remains positive. This feature distinguishes a non-equilibrium system from an equilibrium one with anisotropic interactions of diffusion constants. For $T < T_c$, we must have $r_{\perp} < 0$, to yield the correct steady state solution (which we

denote by ϕ_c) for two-phase coexistence, with the interfaces tangent to E. Since a configuration with interfaces perpendicular to E has never been observed in simulations, it is *sufficient* to assume $r_\parallel > 0$, so that (1) will not admit such solutions. (For our purposes, the necessary condition is weaker. However, the physics associated with it is less clear.)

2. For systems in equilibrium, the fluctuation–dissipation theorem insures that the noise correlations operator, N, be proportional to the diffusion operator, $D \equiv r_\parallel \partial^2 + r_\perp \nabla^2$. An important characteristic of non-equilibrium steady states is the violation of this theorem so that $N \not\propto D$ in general, giving rise to power-law decay of correlations without any parameter tuning, i.e., generic scale invariance (Zhang *et al* 1988, Grinstein *et al* 1990, 1991, Cheng *et al* 1991).

Although in principle a fully non-local and nonlinear equation for the interface should be derived from (1) (Leung 1988, Hernández-Machado and Jasnow 1988), there are some technical, as well as philosophical, difficulties in this approach. Here, we rather focus on the linearized equation and discuss novel aspects in two specific properties: (a) the dispersion relations for capillary waves, which governs the decay rate of long-wavelength fluctuations, and (b) roughening of the interface, via the relationship between the statistical width of the interface and system sizes. For (a), we need only the deterministic part of (1), while for (b), we need to consider the noise also.

Dispersion relation. It is easy to find the stationary solution ϕ_c to (1) under the boundary conditions $\phi \to \pm M$ as $z \to \pm\infty$, where $M = -6g_\perp / r_\perp$ is the magnitude of the spin densities of the two ordered phases, and z labels an arbitrarily chosen direction among the $(d-1)$-dimensional subspace orthogonal to E, since the system is rotationally invariant in that subspace, ϕ_c is simply $M \tanh(z/\xi)$, describing a planar interface of intrinsic width $\xi = (-2/r_\perp)^{1/2}$ centred at $z = 0$, with its normal along z and perpendicular to E. Of the $d-1$ coordinates orthogonal to z, let $x_\parallel$ be parallel to E, and y be the remaining $(d-2)$-dimensional vector orthogonal to both $x_\parallel$ and z. For small fluctuations about ϕ_c, we linearize (the determinisitic part of) (1) in $\chi \equiv \phi - \phi_c \propto e^{iqx_\parallel + ik\cdot y}$, to arrive at

$$\begin{aligned} -i\omega\chi &= \{-r_\parallel q^2 + r_\perp(\partial_z^2 - k^2) - [\alpha q^4 + \beta(\partial_z^2 - k^2)^2 - 2\gamma q^2(\partial_z^2 - k^2)] \\ &\quad -\tfrac{1}{2}[g_\parallel q^2 - g_\perp(\partial_z^2 - k^2)]\phi_c^2\}\chi \\ &\equiv -F\chi. \end{aligned} \tag{2}$$

Thus we have exploited the translational symmetry and partly diagonlized the differential operator in the usual way. Finally, diagonalizing F will give the dispersion relation, $\omega(q, k)$, associated with a particular mode (eigenfunctions of F). To take full advantage of an anticipated small q and k expansion, *the fluctuation operator is decomposed as* $F = AB + \Delta$, with

$$\begin{aligned} A &= \kappa^2 - \partial_z^2 \\ B &= r_\parallel + bq^2 - \beta(\partial_z^2 - k^2) + \tfrac{1}{2}g_\perp \phi_c^2 \\ \Delta &= \varepsilon q^2 + O(q^4) \end{aligned} \tag{3}$$

where $\kappa \equiv (k^2 + g_\parallel q^2/g_\perp)^{1/2}$, $b \equiv 2\gamma - g_\parallel \beta / g_\perp$ and $\varepsilon \equiv r_\parallel - g_\parallel r_\perp / g_\perp$.

We emphasize that, due to anisotropies induced by random stirring, $\varepsilon > 0$. Without these anisotropies, $\Delta = 0$ and the usual form of model B is recovered (Jasnow and Zia

1987). The operator A arises from conservation; its inverse supplies the non-local operator discussed above.

Note that A is a positive operator, B is non-negative and Δ is just a constant, while all three are Hermitian. So, F can be diagonalized: $F|m\rangle = \lambda_m|m\rangle$, with a non-negative real spectrum, though F itself is not Hermitian. Thus, at least locally, our solution is stable. For later convenience, we note that $F^+ = BA + \Delta$ is the adjoint of F, diagonalized by the adjoint eigenstates $|\tilde{m}\rangle$. In our case, they are simple: $|\tilde{m}\rangle = A^{-1}|m\rangle$. The eigenstates can be orthonormalized by $\langle \tilde{m} | n' \rangle = \delta_{mm'}$.

The eigenstates of B are known (Langer 1967), but only one (band) is the result of ϕ_c breaking the translational invariance in z. Therefore, we expect $\phi'_c \equiv d\phi_c/dz$ to remain an exact eigenstate of F with $q = k = 0$. This is the Goldstone mode. By a straightforward perturbation expansion in small q and k, we find the lowest eigenstates associated with it. Labelled by $|1\rangle$, they are, apart from the plane wave factors, $\mathcal{N}\phi'_c + O(q^2)$, where $\mathcal{N}$ is a normalization constant. These are the capillary-wave excitations about a planar interface, with wavevector (q, k). Their dispersion relation is:

$$-i\omega = -\lambda_1 = -\varepsilon q^2 - \frac{2}{3\xi}\kappa(bq^2 + \beta k^2) + \dots. \tag{4}$$

As a result of the first term, the capillary waves can no longer be separated from the bulk diffusive modes by the wavevector dependence in the dispersion relations. Unlike model B where $-i\omega \propto q^3$ (Langer and Turski 1977, Jasnow and Zia 1987), they cannot be singled out as the slowest modes simply by considering fluctuations with the longest wavelengths.

However, note that ε vanishes with the drive, so that, for small driving fields, an interesting crossover from a q^2 behaviour to q^3 can occur in $d = 2$, where $\kappa \propto q$. A similar crossover, between anisotropic scaling, occurs in $d \geq 3$.

Roughening. We now outline the calculation of the height-height correlation function and the statistical width squared, w^2, for the interface. For all the cases we know, analysis linear in χ is sufficient to capture the correct physics, e.g., sizes $(L_\parallel, L_\perp)$ dependence and roughness.

For our purposes, it is more convenient to recast (2) in the form of a dynamic functional, $\mathcal{J}$:

$$\mathcal{J} = \int \tilde{\phi}(-i\omega\chi + F\chi - N\tilde{\phi}) \tag{5}$$

with $\tilde{\phi}$ the Martin-Siggia-Rose response field conjugate to ϕ (Martin *et al* 1973, Janssen 1976, DeDominicis 1976). Since we are considering only the quadratic approximation to $\mathcal{J}$, the correlations for the fluctuations χ are easily found, in matrix form:

$$\langle \chi\chi \rangle = 2(-i\omega + F)^{-1} N (i\omega + F^+)^{-1}. \tag{6}$$

Inserting complete sets into (6), we integrate over ω to give equal-time correlations:

$$\langle \chi_m \chi_{m'} \rangle = \frac{2\langle \tilde{m}|N|\tilde{m}' \rangle}{(\lambda_m + \lambda_{m'})}. \tag{7}$$

This equation is, in fact, completely general, as long as F has a real, positive spectrum. As a comparison, for systems near equilibrium, the fluctuation dissipation theorem insures that $F = N\Gamma$, where Γ is the 'propagator' in the static theory, similar in form to B in (3). Then, simple manipulation using (6) leads to the equal-time correlation Γ^{-1}. In our case, $N = n_\perp A + O(q^2)$, so that (7) also simplifies to $\langle \chi_m \chi_{m'} \rangle = n_\perp \delta_{mm'}/\lambda_m$, at the lowest order.

Now, we are ready to find the height-height correlation function C, associated with capillary waves, described by $\chi = h(x)\phi_c'$. Recalling that $|1\rangle \propto \phi_c'$, we have

$$C(q,k) = \frac{n_\perp \mathcal{N}^2}{\lambda_1}[1 + O(q^2)]$$

$$\approx \frac{n_\perp \kappa}{2M^2[\varepsilon q^2 + 2\kappa(bq^2 + \beta k^2)/3\xi]} \tag{8}$$

where we have used the fact that ϕ_c' is essentially a constant $(2M)$ in momentum space for evaluating $\mathcal{N}$.

In $d = 2$, $\kappa \propto q$, so that $C(q) - 1/q$ for small q, in contrast to the well known $1/q^2$ for equilibrium. For $d \geqslant 3$, correlations are highly anisotropic, being the usual $1/k^2$ in directions transverse to the field.

Finally, the width squared is given by

$$w^2 = \int_{1/L_\perp} d^{d-2}k \int_{1/L_\parallel} dq\, C(q,k). \tag{9}$$

Completing the integrations, we get $\ln L_\parallel$ behaviour in $d = 2$ (for $L_\parallel \gg 1/\varepsilon\xi$), and finite w^2 in $d = 3$, for both $L_\parallel$ and $L_\perp$ going to infinity in either order. This is in sharp contrast to the interfaces of equilibrium systems, which are rough for $d \leqslant 3$ (van Beijeren and Nolden 1987).

At present, there is not yet any experimental result, but the system can easily be simulated on a computer. Preliminary results indeed show very strong suppression of roughening, though it is not yet possible to assert from the data the predicted Ln L behaviour in 2D. One problem which plagues the data analysis is unusually strong finite-size effects. In equilibrium systems, the finite-size dependence of $w^2(L_\parallel, L_\perp)$ is completely dominated by $L_\parallel$, since the effects of finite $L_\perp$ are exponentially small—a consequence of exponential decay of bulk correlation well below T_c. In contrast, bulk correlation in this class of non-equilibrium steady states is known to decay as $1/x^d$ (Zhang *et al* 1988, Cheng *et al* 1991). Thus, we can expect the presence of long-range effect which, mediated through long-range correlations from the boundaries to the interface, vanishes only as a power of $L_\perp$.

We have indeed observed such a finite-size effect in simulation, and we can also demonstrate it analytically. For simplicity, consider $d = 2$. Replacing the lower limit of integration in (8) by $1/L_\perp$, we deduce that $C(q)^{-1} \sim \varepsilon(q - a_1/L_\perp + \ldots)$ for $q \gg 1/L_\perp$ under periodic boundary conditions, with a_1 of order unity. Consequently, careful extrapolation for both longitudinal *and* transverse system sizes are necessary when simulating roughening in steady states.

Before we conclude, we comment on the generality of our results. One obvious question is whether the ϕ^3 terms in (1) (with origins in a ϕ^4 potential) are special. Looking over our analysis, it is straightforward to see that our results will be valid for *arbitrary* nonlinear terms, provided those associated with ∂^2 and ∇^2 are proportional to each other. We believe, though we have not proved, that the situation is less restrictive. Work is under way on exactly solvable models, from which we hope to gain enough insight to prove the more general case.

To conclude, we studied interfacial properties in a non-equilibrium steady state system. We emphasized the need of working with the bulk equation of motion in the two-phase region. Extracting those modes which have a Goldstone character, we identify interface fluctuations and showed that, unlike in equilibrium, 'static' properties cannot be studied without considerations for the full dynamics (see discussion after (7)). For the specific model of randomly driven diffusive systems, we calculated the dispersion relation, the correlation function, and the statistical width of the interface. Analytical evidence of roughening suppression, reminiscent of our previous finding by simulations in the uniformly driven system, is provided for the first time. We believe that this study will serve as an excellent starting point for understanding interfacial behaviour in more complex non-equilibrium systems.

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